ACCURATE PREDICTION OF VIBRONIC LEVELS AND BRANCHING RATIOS FOR LASER-COOLABLE LINEAR POLYATOMIC MOLECULES: APPLICATIONS TO CAOH, SROH, AND YBOH

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We report calculations of vibronic levels and branching ratios for laser-coolable linear polyatomic molecules to an accuracy and completeness to be useful to guide experimental studies. The present computational scheme consists of a multi-state quasidiabatic Hamiltonian with relevant perturbations such as Renner-Teller, linear vibronic, and spin-orbit coupling, coupled-cluster calculations for adiabatic potential energy surfaces and molecular parameters, and discrete variable representation calculations for vibronic levels and wave functions. The computed levels and branching ratios for the $A^2\Pi_{1/2} \rightarrow X^2\Sigma_{1/2}$ transitions of CaOH, SrOH, and YbOH show promising agreement with experimental measurements. Based on the computed branching ratios, laser-cooling SrOH requires fewer repumping lasers than CaOH. The calculations also elucidate intensity-borrowing mechanisms for nominally symmetry-forbidden transitions. A close inspection of computational results further reveals it beneficial to avoid Fermi resonances in designing laser-coolable molecules.